Evidence for a two component magnetic response in UPt_3

A. Yaouanc,[1] P. Dalmas de Réotier,[1] F.N. Gygax,[2] A. Schenck,[2] A. Amato,[3] C. Baines,[3] P.C.M. Gubbens,[4] C.T. Kaiser,[4] A. de Visser,[5] R.J. Keizer,[5] A. Huxley[1] and A.A. Menovsky[5]

[1] *Commissariat à l'Energie Atomique, Département de Recherche Fondamentale sur la Matière Condensée F-38054 Grenoble Cedex 9, France*

[2] *Institute for Particle Physics, Eidgnössische Technische Hochschule Zurich, CH-5232 Villigen PSI, Switzerland*

[3] *Laboratory for Muon Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland*

[4] *Interfacultair Reactor Instituut, Delft University of Technology, 2629 JB Delft, The Netherlands*

[5] *Van der Waals-Zeeman Instituut, Universiteit van Amsterdam, 1018 XE Amsterdam, The Netherlands*

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The magnetic response of the heavy fermion superconductor UPt_3 has been investigated on a microscopic scale by muon Knight shift studies. Two distinct and isotropic Knight shifts have been found for the field in the basal plane. While the volume fractions associated with the two Knight shifts are approximately equal at low and high temperatures, they show a dramatic and opposite temperature dependence around T_N. Our results are independent on the precise muon localization site. We conclude that UPt_3 is characterized by a two component magnetic response.

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The hexagonal heavy fermion superconductor UPt_3 is attracting much interest because it has been established as an unconventional superconductor as seen by the existence of three distinct superconducting phases in the magnetic field-temperature plane [1,2]. In zero-field the two superconducting phase transitions occur at ~ 0.475 K and ~ 0.520 K. It is usually thought that this complex phase diagram arises from the lifting of the degeneracy of a multicomponent superconducting order parameter.

The most popular candidate for such a symmetry-breaking field is the short range antiferromagnetic order characterized by a Néel temperature of $T_N \simeq 6$ K and an extremely small ordered magnetic moment (0.02 (1) μ_B/U-atom in the limit $T \to 0$ K) oriented along the a^* axis ($\equiv b$ axis). The magnetic order has only been observed by neutron [3] and magnetic x-ray [4] diffractions.

Nuclear magnetic resonance [5] and zero-field muon spin relaxation [6] measurements as well as macroscopic studies have failed to prove the existence of static antiferromagnetic order on high quality samples [7]. Here we present transverse high-field muon spin rotation (μSR) data which present anomalies around T_N. Moreover, they show that UPt_3 is characterized by a two component magnetic response at least up to 115 K.

In the transverse μSR technique [8], polarized muons are implanted into a sample where their spins $\mathbf{S}_\mu$ ($S_\mu = 1/2$) precess in the local magnetic field $\mathbf{B}_{\rm loc}$ until they decay. The sample is polarized by a magnetic field $\mathbf{B}_{\rm ext}$ applied perpendicularly to $\mathbf{S}_\mu(t=0)$. $\mathbf{S}_\mu(t)$ is monitored through the decay positron. By collecting several million positrons, one can readily obtain an accurate value for the field at the muon site(s).

We present results for three samples. Two samples have been grown in Grenoble. Each consists of crystals glued on a silver backing plate and put together to form a disk. They differ by the orientation of the crystal axes relative to the normal to the sample plane: either the a^* or c axis is parallel to that direction. Measurements have therefore been carried out either with $\mathbf{B}_{\rm ext}$ parallel to a^* or c. The third sample has been prepared in Amsterdam. It is a cube of $5 \times 5 \times 5$ mm^3 which has been glued to a thin silver rod. The measurements on this sample have been done only with $\mathbf{B}_{\rm ext} \parallel a$. The Grenoble samples have already been used for zero-field [6] and transverse low-field [9] μSR measurements. Their high quality is demonstrated by the splitting of the two zero-field superconducting transitions as seen by specific heat [9] and the low residual resistivities which are among the lowest ever reported ($\rho_c(0) = 0.17$ $\mu\Omega$ cm and $\rho_{a^*}(0) = 0.54$ $\mu\Omega$ cm [10]). The Amsterdam sample is of a somewhat lesser quality in terms of the residual resistivities which are roughly a factor 3 higher than for the Grenoble sample. Nevertheless the double superconducting transition is clearly resolved in the specific heat.

The measurements have been performed at the low temperature facility (LTF) and at the general purpose spectrometer (GPS) of the μSR facility located at the Paul Scherrer Institute. The LTF spectra have been obtained for temperatures between 0.05 K and 10 K and $B_{\rm ext}$ of 2.3 T (only for two measurements), 2 T and 1.5 T. The GPS data have been taken with $B_{\rm ext} = 0.6$ T for $1.7 \text{ K} \leq T \leq 200$ K. A high statistic GPS measurement has been carried out at 50 K with $B_{\rm ext} = 0.45$ T. The GPS measurements have been performed with an electrostatic kicker device on the beam line which ensures that only one muon at a time is present in the sample [11]. With such a device, the signal to noise ratio is strongly enhanced and the time window is extended to ~ 18 μs. For both spectrometers $\mathbf{B}_{\rm ext}$ has been applied along the muon beam direction and a spin rotator has been used to flip the muon spin away from the muon momentum.

We expect to observe a sum of oscillating signals, each corresponding to a given type of muon environment. An

extra signal originating from muons stopped in the sample surroundings, basically a silver backing plate, is also expected.

In Fig. 1 we present two Fourier transforms of spectra. Two lines from the sample are clearly detected for $\mathbf{B}_{\rm ext} \parallel a^*$. A symmetric single line is observed for $\mathbf{B}_{\rm ext} \parallel c$. For the whole temperature range investigated two components are found for $\mathbf{B}_{\rm ext} \perp c$ and only a single component is detected for $\mathbf{B}_{\rm ext} \parallel c$. In Fig. 2 we present a time spectrum which clearly shows the existence of the two components far into the paramagnetic regime for $\mathbf{B}_{\rm ext} \perp c$.

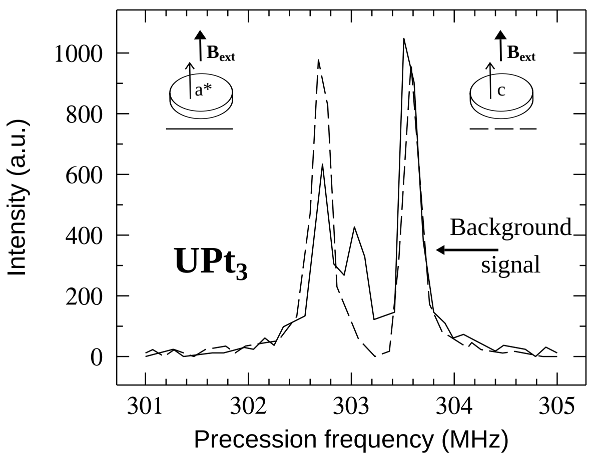

FIG. 1. Two Fourier transforms of spectra recorded at 2.6 K with $B_{\rm ext} = 2.3$ T. $\mathbf{B}_{\rm ext}$ is either parallel to the a^* or c axis. The background signal was intentionally enlarged in this measurement to evidence the difference between the applied field and the field at the muon site. This was achieved by fixing a 10 mm diameter Ag mask on the sample, resulting in a reduced effective sample size. The line at ~ 303.6 MHz originates from the Ag mask.

We first discuss the spectra recorded for $\mathbf{B}_{\rm ext} \perp c$ which have been analyzed with the polarization function $P_X(t)$ written as the sum of three components:

$$aP_X(t)= a_{\rm F}\cos(\omega_{\rm F}t)\exp\left(-\Delta^2t^2/2\right) \\ +a_{\rm S}\cos(\omega_{\rm S}t)\exp(-\lambda t) + a_{\rm bg}\cos(\omega_{\rm bg}t)\exp(-\lambda_{\rm bg}t). \quad (1)$$

The first two components describe the μSR signal from the sample and the third accounts for the muons stopped in the background. The subscripts F and S refer to the first and second components, respectively. a_α is the initial asymmetry of component α oscillating at the pulsation frequency $\omega_\alpha = 2\pi\nu_{\mu,\alpha} = \gamma_\mu B_{{\rm loc},\alpha}$ where $\nu_{\mu,\alpha}$ is the precession frequency of component α and γ_μ the muon gyromagnetic ratio ($\gamma_\mu = 851.6$ Mrad s^{-1}T^{-1}). a_α is proportionnal to the fraction of muons experiencing field $B_{{\rm loc},\alpha}$. The envelop of the first component is best fitted by a Gaussian function, while the envelop of the second component is better described by an exponential damping. We stress that the measured temperature dependences of the two initial asymmetries and frequencies are not influenced by the choice of the envelop functions.

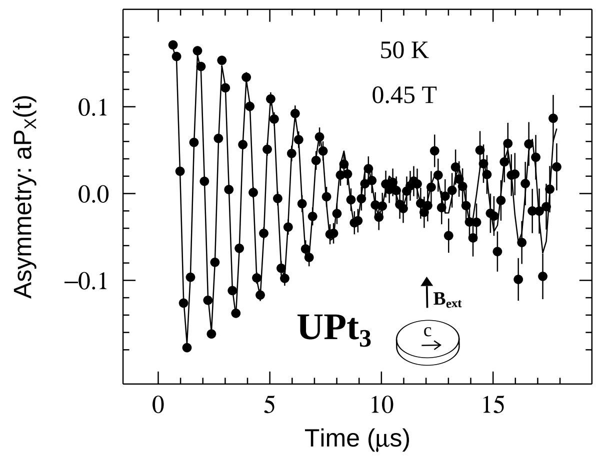

FIG. 2. A spectrum recorded at 50 K in a field of 0.45 T applied perpendicular to the c axis of UPt$_3$ and presented in a frame rotating at a precession frequency of 60 MHz. This experiment was perfomed in a setup designed to get the smallest possible background ($a_{\rm bg} \simeq 0.017$). The solid line is a fit to a sum of three oscillating components: two from the sample and one from the background. The reduced asymmetry observed on the plot around 10 μs, which is too small to be explained by a beating of the UPt$_3$ and background signals, reflects the beating of the two signals originating from UPt$_3$.

Δ is approximately independent of the temperature and amounts to $\simeq 0.55$ MHz at high field. It roughly scales with $B_{\rm ext}$. λ is independent of $B_{\rm ext}$ and is equal to $\lambda \simeq 0.14$ MHz at the lowest temperature. It decreases when the temperature is increased and becomes so small above 4 K that it can be fixed to zero. The values of the damping rates may reflect only partially the intrinsic properties of UPt$_3$ because of the field inhomogeneity due to the demagnetization field. However, Δ and λ are remarkably small, indicating that the magnetic inhomogeneity detected for the two components is small.

In Fig. 3 we display the temperature dependence of the two initial asymmetries and the associated relative frequency shifts, K_μ. These plots concern the spectra taken with $\mathbf{B}_{\rm ext} \perp c$ and for $T \leq 14.7$ K. K_μ, which is the local magnetic susceptibility at the muon site, is usually called the Knight shift. It is deduced from the measured relative frequency shift, $K_{\rm exp}$, after correcting for the Lorentz and demagnetization fields. $K_{\rm exp}$ is defined by $K_{\rm exp} = \mathbf{B}_{\rm ext}\cdot(\mathbf{B}_{\rm loc} - \mathbf{B}_{\rm ext})/B^2_{\rm ext}$. We have determined $B_{\rm ext}$ with a gaussmeter or through the pulsation frequency of the background: $B_{\rm ext} = \omega_{\rm bg}/\gamma_\mu$. Since the Knight shift of the background is very small (K_μ for silver is $\simeq 94$ ppm [12]), this is a very good approximation. Although the Lorentz and demagnetization correction modifies substantially the absolute value of the Knight shift, qualitatively it does not influence its temperature dependence. The conclusions we shall draw from our data are independent of the uncertainty related to the correction.

The results of Fig. 3 show that the Grenoble and Amsterdam samples yield consistent results. Since, as indicated in the figure, the measurements have been done either with $\mathbf{B}_{\mathrm{ext}} \parallel a^*$ or $\mathbf{B}_{\mathrm{ext}} \parallel a$, we conclude that the μSR response is isotropic in the basal plane. The data display also two remarkable features.

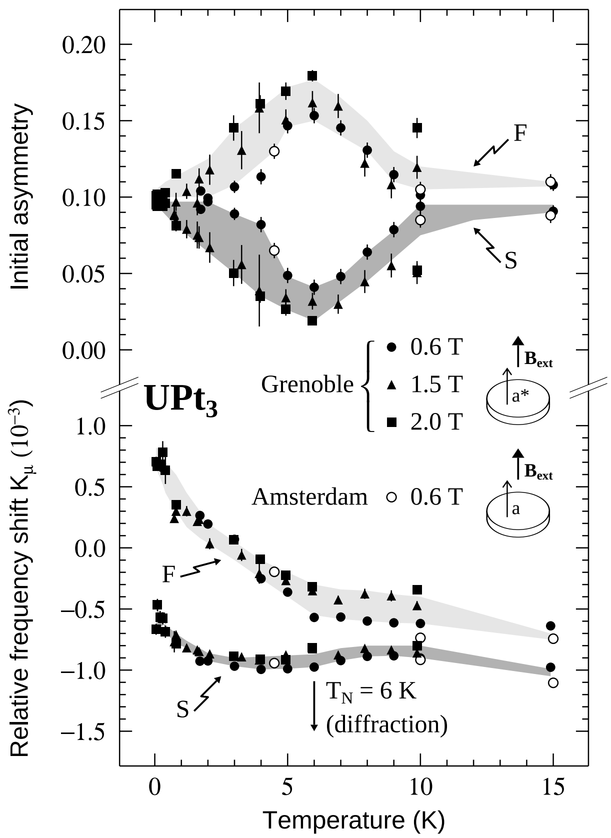

FIG. 3. Temperature dependence of the initial asymmetries and Knight shifts K_μ with $\mathbf{B}_{\mathrm{ext}}$ perpendicular to the c axis. The data are for 0.5 K $\leq T <$ 15 K, three field intensities and two samples denoted as Grenoble and Amsterdam. K_μ is corrected for Lorentz and demagnetization fields. The actual value of K_μ is subject to an uncertainty due to the demagnetization correction. Nevertheless the shape of $K_\mu(T)$ is independent on this correction. The F and S letters denotes the two components.

First, the frequency splitting between the two lines is relatively large at low T, decreases rapidly as T increases up to $\sim$ 6 K and exhibits a shallow minimum around 10 K. It increases again for higher temperatures (not shown). This explains the possibility of observing the beating between the two oscillating components at 50 K as shown in Fig. 2. Only the F shift has a strong temperature dependence below 6 K while the S shift is practically temperature independent down to 0.4 K, below which its absolute value slightly decreases. Since 6 K is the T_N value as determined by neutron diffraction on our samples, the temperature dependence of the F shift provides a signature of the Néel temperature.

The second feature is probably the most striking: we observe two muon precession frequencies with approximately equal initial asymmetries in the whole temperature range (0.05 K $\leq T \leq$ 200 K, the region $T >$ 15 K is not shown in Fig. 3) except near T_N ($T_N \pm$ 4 K) where a_{F} increases at the expense of a_{S}.

In this temperature range a trend for a larger difference between these initial asymmetries seems to be present at high field. However this trend might not be meaningful since the signal to noise ratio for the 2.0 T and 1.5 T spectra is not as good as for the 0.6 T spectra. An eventual field effect on a_{F} and a_{S} could only be confirmed by measurements with the electrostatic kicker device at all fields. Since high-field neutron diffraction [13,14] did not detect any sizeable change in the relative population of the three equivalent antiferromnagnetic domains we do not expect a field effect on the initial asymmetries.

The spectra recorded with $\mathbf{B}_{\mathrm{ext}} \parallel c$ have been analysed with a formula similar to Eq. 1 with $a_{\mathrm{S}} = 0$. The precession frequency varies smoothly in temperature. The Gaussian damping rate scales again with B_{ext} ($\sim$ 0.42 MHz at 1.5 T) and is essentially temperature independent up to $\sim$ 30 K above which temperature it drops smoothly to very small values.

In Fig. 4 we present the K_μ data recorded for $B_{\mathrm{ext}} =$ 0.6 T with 1.7 K $\leq T \leq$ 115 K as a function of the bulk susceptibility χ_B. This is a so called Clogston-Jaccarino plot, the temperature is an implicit parameter. The bulk susceptibilities for the different orientations have been measured on the Grenoble samples and are similar to those of Ref. [15]. Classically, we should find K_μ scaling with the susceptibility. This is approximately observed for $\mathbf{B}_{\mathrm{ext}} \parallel c$ but not for $\mathbf{B}_{\mathrm{ext}} \perp c$. In addition, as already pointed out when discussing $K_\mu(T)$, the Clogston-Jaccarino plots clearly show that while the F Knight shift provides a signature of T_N, such a signature is absent for the S Knight shift. The data of Fig. 4 suggest that K_μ passes smoothly through T_N for $\mathbf{B}_{\mathrm{ext}} \parallel c$, although the almost constant value of $\chi_B(T)$ at low temperature does not allow for a definite statement.

We now discuss the muon diffusion properties and localization site in UPt$_3$. The shape of the zero-field depolarization and the constant value of the related damping rate show that the muons are static and occupy the same site in the muon time scale, at least below 30 K [6]. Our transverse field measurements suggest that in fact the muon is diffusing only above 115 K because the frequency splitting collapses above that temperature (not shown). Since we focus on the properties of UPt$_3$ itself, we only consider the data for which the muon is static. Thus the anomalous temperature dependence of the two initial asymmetries around T_N for $\mathbf{B}_{\mathrm{ext}} \perp c$ can not be due to muon diffusion. The eventual existence of two distinct muon sites can not explain our data since their relative occupancy should not change for a static muon. Interestingly, the analysis for U(Pt$_{0.95}$Pd$_{0.05}$)$_3$ of the an-

gular dependence of K_μ shows that the muon occupies only one site, the 2a site in Wyckoff notation (P6_3/mmc space group) in this related compound [16].

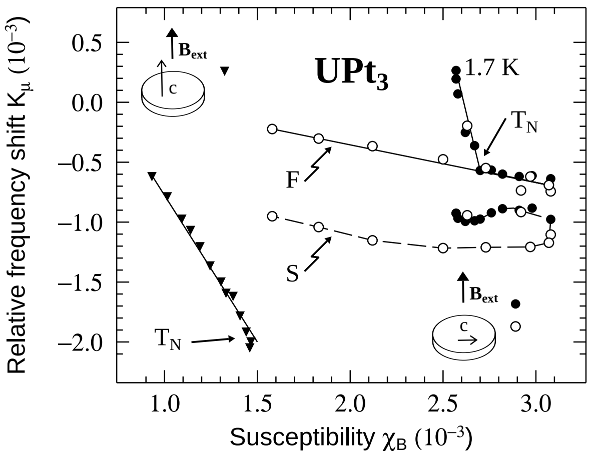

FIG. 4. Clogston-Jaccarino plots obtained for $B_{\rm ext}$ = 0.6 T. For $\mathbf{B}_{\rm ext} \perp c$ we use the same symbol convention as in Fig. 3. The filled triangles correspond to $\mathbf{B}_{\rm ext} \parallel c$. The temperature is an implicit parameter: 1.7 K $\leq T \leq$ 115 K. K_μ is extracted from the reported μSR measurements and the bulk susceptibility χ_B from measurements on our samples. The lines are guides to the eyes. Note the drastic change of slope at T_N for the F set of data.

Our results are understood if we suppose that the muon occupies only one magnetic site and the sample is intrinsically inhomogeneous: it consists of two regions with slightly different magnetic responses and relative volumes which are temperature dependent. While near T_N one region dominates, outside that temperature range the two regions occupy approximately equal volumes. An alternative explanation for our data could be the existence of a complex magnetic structure leading to the observed μSR response. However this would imply a more involved magnetic structure than the one published [3,4,13,14]. In addition it is difficult to imagine that a magnetic structure can influence the muon response up to at least 20 T_N. Therefore we disregard this latter explanation.

The facts that the magnetic phase transition is only detected by transverse high-field and not by zero-field μSR measurements [6,17] are not inconsistent. It is not unexpected to observe below T_N a new source of quasi-static magnetic polarization induced by the applied field which leads to an extra Knight shift.

Bulk magnetic susceptibility does not detect the phase transition since the relative sensitivity in these conditions is $\simeq 10^{-3}$. As shown in Fig. 3, this is not enough.

The results obtained by the μSR and magnetic diffraction techniques are not contradictory. The diffraction results simply mean that the difference in the scattering properties of the two regions may be too subtle to be distinguished.

We now consider the possible origin for the additional Knight shift observed below T_N for the F component. A change of the magnitude of the moments is excluded since high-field neutron diffraction measurements do not detect any sizeable influence of a field up to 12 T [13,14]. Two mechanisms producing an additional shift can be imagined. The first mechanism involves the dipolar field produced at the muon site by the ordered uranium moments. A rotation of these moments induced by the applied field leads to an additionnal field at the muon site. A small rotation is not excluded by neutron diffraction since this technique gives an upper bound rotation angle as large as 26° [13]. But it is surprising, for a magnet with moments oriented along the a^* direction to observe the same K_μ for $\mathbf{B}_{\rm ext} \parallel a$ and $\mathbf{B}_{\rm ext} \parallel a^*$ (see Fig. 3). The second possible origin focuses on the itinerant character of the magnetism of UPt$_3$. In this picture the additional shift is a measure of the enhancement of the magnetic susceptibility of the conduction electrons below T_N. UPt$_3$ being a planar magnet with a negligible planar anisotropy, the enhancement should be isotropic in the plane perpendicular to c and no enhancement should be observed for $\mathbf{B}_{\rm ext} \parallel c$. This is consistent with our data.

Our most surprising result is the existence of the two components when $\mathbf{B}_{\rm ext} \perp c$. Since the associated damping rates are small, we infer that the magnetic disorder is small. The near equality in most of the temperature range of the two initial asymmetries suggests that the two regions originate from a periodic modulation. The behaviour of the initial asymmetries near T_N implies that the proposed modulation is strongly coupled to the magnetic order parameter. The structural modulation observed by electron microscopy and diffraction some years ago [18] might be related to the regions discussed here. However it has never been seen thereafter including in our samples.

In summary we have discovered by transverse high-field μSR measurements the existence of a two component magnetic response. While the volume fraction associated with these components is equal below $\sim$ 2 K and above $\sim$ 10 K, it is strongly temperature dependent around T_N. We also observe a signature of the magnetic transition for one of the two components. Our results are naturally explained if UPt$_3$ is intrinsically inhomogeneous at least in a applied field.

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